

Lateral Chirality-sorting Optical Spin Forces in Evanescent Fields

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(Dated: December 23, 2014)

The transverse component of the spin angular momentum of evanescent waves gives rise to lateral optical forces on chiral particles, which have the unusual property of acting in a direction in which there is neither a field gradient nor wave propagation. As their direction and strength depends on the chiral polarizability of the particle, they act as chirality-sorting and may offer a mechanism for passive chirality spectroscopy. The absolute strength of the forces also substantially exceeds that of other recently predicted sideways optical forces, such that they may more readily offer an experimental confirmation of the phenomenon.

PACS numbers: 12345

INTRODUCTION

Chirality (from Greek $\chi\epsilon\rho$ (*kheir*), 'hand') is a type of asymmetry where a geometric object can not be made to coincide with its mirror image through any proper spatial rotation [1]. While they share all properties other than their helicity, a chiral object and its mirror image differ in their interaction with a chiral environment, such as biological systems. The analysis of and separation by chirality of substances consequently represents a highly important challenge in research and industry, affecting especially pharmaceuticals and agrochemicals [2–5]. While the sorting of substances by chirality normally has to be addressed through the introduction of a specific chiral resolving agent [6], the manifestation of chirality in the electromagnetic response of materials has raised the question of passive sorting using optical forces [7–12].

In Ref. [12] Wang et. al. recently predicted an electromagnetic plane wave to exert a lateral optical force on a chiral particle above a reflective surface, which emerges as the particle interacts with the reflection of its scattered field. This highly unusual force acts in a direction in which there is neither wave propagation nor an intensity gradient, and deflects particles with opposite helicities towards opposite sides. Apart from its fundamental interest, such a force may in theory be useful for all-optical enantiomer sorting with a single, unstructured beam. In Ref. [13], Bliokh et. al. almost simultaneously predicted another lateral force that is exerted on non-chiral particles in an evanescent wave. This force is a consequence of the linear momentum of the wave that is associated with its spin angular momentum (SAM), which was first described by F. J. Belifante in the context of quantum field theory, and which vanishes for a propagating plane wave [14, 15].

In this letter we investigate a third mechanism, in which a force emerges as the spin angular momentum itself gives rise to a linear momentum transfer as it inter-

acts with chiral materials. This effect enables the tailoring of chirality-sorting optical forces by engineering the local SAM density of optical fields, such that fields with transverse SAM can be used to achieve lateral forces. We calculate the forces emerging in an evanescent field with transverse SAM, and show that the magnitude of the lateral force substantially exceeds those of the previously predicted lateral forces. In the following section of this

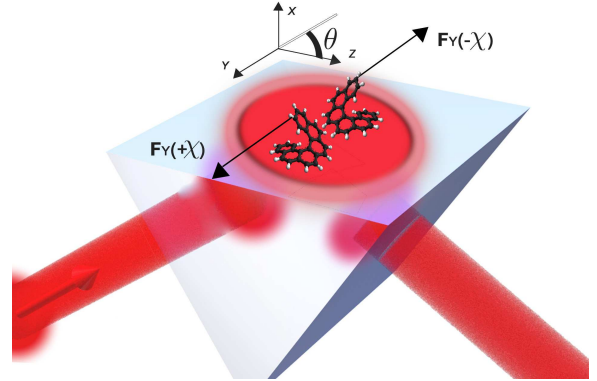


FIG. 1. Chirality-dependent Lateral Forces in an Evanescent Field: An evanescent field arises as light in a high index medium (n_1) is totally internally reflected at the interface with a low index medium (n_2) at an angle θ beyond the critical angle. Particles in an evanescent field with transverse spin angular momentum experience lateral forces depending on their chiral polarizability χ , with particles with opposite helicities experiencing lateral forces in opposite directions.

paper we provide a brief general discussion of the optical forces exerted by an electromagnetic wave on a small chiral particle and treat the specific case of an evanescent wave. We then calculate the lateral force on a nano sphere, and compare it to the strength of the lateral force that may arise due to the other two effects discussed by Wang et. al and Bliokh et. al [12, 13].

OPTICAL FORCES ON SMALL PARTICLES IN AN EVANESCENT FIELD

The optical forces exerted on an object is rigorously calculated by first finding the distribution of electromagnetic fields and then integrating the Maxwell stress tensor over a surface enclosing the object. The scattering problem associated with finding the field distributions is tremendously simplified in the dipole approximation, which holds in the so-called Rayleigh limit that applies to particles much smaller than the wavelength. The dipole solution is sufficiently accurate for most molecular scattering problems and is furthermore the basis for efficient numerical methods that are used to find solutions for complex larger objects [16]. We consider the optical forces on a small particle in a source-free, lossless, non-dispersive and isotropic medium with relative electric permittivity ϵ and relative magnetic permeability μ . In the dipole approximation, the time-averaged total optical force \mathbf{F} exerted on the particle by a monochromatic electromagnetic wave is given by [17]:

$$\mathbf{F} = \underbrace{\frac{1}{2}\text{Re}\{\mathbf{d}(\nabla \otimes \mathbf{E}^*) + \mathbf{m}(\nabla \otimes \mathbf{H}^*)\}}_{\mathbf{F}_0} - \underbrace{\frac{k^4}{3}\text{Re}\left\{\sqrt{\frac{1}{\mu\epsilon}}(\mathbf{d} \times \mathbf{m}^*)\right\}}_{\mathbf{F}_{\text{int}}}, \quad (1)$$

where \mathbf{E} and \mathbf{H} are the electric and magnetic field vectors of the incident electromagnetic wave at the location of the particle; \mathbf{d} and \mathbf{m} are the electric and magnetic dipole moments of the particle; $k = n\omega/c$ is the wavenumber of the electromagnetic wave; ω is the frequency; c is the speed of light in vacuum; and $n = \sqrt{\epsilon\mu}$ the refractive index of the medium. The fields are written in complex phasor notation throughout this paper, where the factor $\exp(-i\omega t)$ giving the time dependence is implied, and the superscript $*$ denotes the complex conjugate. Vector quantities are indicated by bold letters, and all expressions are given in Gaussian units unless stated otherwise. $\hat{\mathbf{x}}, \hat{\mathbf{y}}$ and $\hat{\mathbf{z}}$ are unit vectors along

the corresponding coordinate axes. The symbol \otimes denotes the dyadic product, so that the terms of the form $\mathbf{W}(\nabla \otimes \mathbf{V})$ in eqn. 1 have elements $[\mathbf{W}(\nabla \otimes \mathbf{V})]_i = \sum_j W_j \partial_i V_j$ for $i, j \in \{x, y, z\}$ [17] and can be written as $\mathbf{W}(\nabla \otimes \mathbf{V}) = (\mathbf{W} \cdot \nabla) \mathbf{V} + \mathbf{W} \times (\nabla \times \mathbf{V})$ in terms of more commonly used vector operators. Equation 1 shows that the optical force has a component \mathbf{F}_0 corresponding to the force exerted by the incident field on the particle's electric and magnetic dipole moments, and a component \mathbf{F}_{int} that results from a direct interaction of the two dipole moments. In case of the evanescent field, it is the \mathbf{F}_{int} term that gives rise to the lateral electromagnetic spin force. Chirality manifests itself in the electromagnetic response of a material through a cross-coupling of the induced electric and magnetic polarizations, such that an electric field also gives rise to a magnetic polarization and vice versa. Indeed, the optical response of chiral materials may be modeled intuitively by a simple coupled oscillator model [18]. Assuming isotropic polarizabilities and no permanent dipole moment, the electric and magnetic dipole moments induced by fields incident on a chiral particle are $\mathbf{d} = \alpha_e \mathbf{E} + i\chi \mathbf{H}$ and $\mathbf{m} = \alpha_m \mathbf{H} - i\chi \mathbf{E}$, where α_e and α_m are the electric and magnetic polarizabilities [12]. The chiral polarizability of the particle χ captures the chiral nature of the dipole, such that setting $\chi = 0$ recovers the case of an achiral dipole. It is worth mentioning that, while the chiral polarizability is an inherently dynamic quantity, the dynamic dipole polarizabilities α_e and α_m differ from the more frequently listed corresponding static polarizabilities $\alpha_e^{(0)}$ and $\alpha_m^{(0)}$ by a radiation correction first derived by Draine as $\alpha_e = \left[1 - (i2k^3/3\epsilon) \alpha_e^{(0)}\right]^{-1} \alpha_e^{(0)}$ and $\alpha_m = \left[1 - (i2k^3/3\mu) \alpha_m^{(0)}\right]^{-1} \alpha_m^{(0)}$ [19]. While the static polarizability is sufficiently accurate in the particular case of the small gold sphere considered in ref. [13], omission of the radiation correction can in general lead to a substantial underestimation of the optical forces [20]. Inserting the expressions for the electric and magnetic dipole moments into equation 1 gives the force on a chiral dipole in an electromagnetic field:

$$\mathbf{F}_0 = \underbrace{\frac{1}{g} \left(\epsilon^{-1} \text{Re}\{\alpha_e\} \nabla u_e + \mu^{-1} \text{Re}\{\alpha_m\} \nabla u_m \right) - \frac{\omega}{g} \text{Re}\{\chi\} \nabla h}_{\text{Gradient Force}} + \underbrace{\frac{2\omega}{g} (\mu \text{Im}\{\alpha_e\} \mathbf{p}_e^o + \epsilon \text{Im}\{\alpha_m\} \mathbf{p}_m^o) - \frac{c}{g} \text{Im}\{\chi\} [\nabla \times \mathbf{p} - 2k^2 \mathbf{s}]}_{\text{Radiation Pressure}} \quad (2)$$

$$\mathbf{F}_{\text{int}} = -\frac{2\omega}{g} \frac{k^3}{3} (\text{Re}\{\alpha_e \alpha_m^*\} \mathbf{p} - \text{Im}\{\alpha_e \alpha_m^*\} \mathbf{p}'' + |\chi|^2 \mathbf{p}) - \frac{2\omega}{g} \frac{2k^4}{3n} (\epsilon \text{Re}\{\chi \alpha_m^*\} \mathbf{s}_m + \mu \text{Re}\{\chi \alpha_e^*\} \mathbf{s}_e). \quad (3)$$

Here we marked the terms corresponding to the familiar gradient force and radiation pressure, both of which

are modified in the presence of chirality. The final term

of eqn. 3 shows that the spin momentum density of the field gives rise to a linear momentum transfer on chiral particles, which occurs in opposite directions for opposite signs of χ , i.e. opposite helicities. The prefactor $g = (4\pi)^{-1}$ arises from the usage of Gaussian units, $u_e = (g\epsilon/4)|\mathbf{E}|^2$ and $u_m = (g\mu/4)|\mathbf{H}|^2$ are the energy densities of the electric and the magnetic field, and $h = (g/2\omega)\text{Im}\{\mathbf{E} \cdot \mathbf{H}^*\}$ is the time-averaged optical helicity density [13, 14]. The time-averaged Poynting momentum density $\mathbf{p} = (g/2c)\text{Re}\{\mathbf{E} \times \mathbf{H}^*\}$ can be decomposed as $\mathbf{p} = \mathbf{p}^o + \mathbf{p}^s$ into a component related to the orbital angular momentum, \mathbf{p}^o , and the Belifante momentum \mathbf{p}^s related to the spin momentum density \mathbf{s} . These quantities can be individually further separated into their electric and magnetic components, in particular $\mathbf{p}^o = \mathbf{p}_e^o + \mathbf{p}_m^o$ and $\mathbf{s} = \mathbf{s}_e + \mathbf{s}_m$ (the supplementary material [20] contains a full list of the definitions of the field quantities, which are discussed in detail in ref. [13]). Invoking a fluid mechanics analogy, the term $\nabla \times \mathbf{p}$ may be interpreted as the vorticity of the Poynting vector flow. $p'' = (g/2c)\text{Im}\{\mathbf{E} \times \mathbf{H}^*\}$ is the imaginary Poynting momentum [13, 21]. For a monochromatic wave, the momentum and spin densities are [13]:

$$\mathbf{p}^s = -\frac{g}{8\omega}\nabla \times \left[(i\mu)^{-1}\mathbf{E} \times \mathbf{E}^* + (i\epsilon)^{-1}\mathbf{H} \times \mathbf{H}^* \right] \quad (4)$$

$$\mathbf{p}^o = -\frac{g}{4\omega}\text{Im}\{\mu^{-1}\mathbf{E}(\nabla \otimes \mathbf{E}^*) + \epsilon^{-1}\mathbf{H}(\nabla \otimes \mathbf{H}^*)\} \quad (5)$$

$$\mathbf{s} = -\frac{g}{4\omega}((\mu i)^{-1}\mathbf{E} \times \mathbf{E}^* + (\epsilon i)^{-1}\mathbf{H} \times \mathbf{H}^*), \quad (6)$$

where the magnetic and electric contributions correspond to the terms proportional to the electric and magnetic fields. Evaluating equations 2-3 for a plane wave $\mathbf{E} = \sqrt{\mu}\mathbf{E}_0\exp(ikz)$ propagating in the $\hat{\mathbf{z}}$ -direction, shows that the forces are entirely in the direction of propagation:

$$\mathbf{F}_0 = [(\text{Im}\{\mu\alpha_e\} + \text{Im}\{\epsilon\alpha_m\})/2 + \text{Im}\{\chi\}n\sigma]Ik\hat{\mathbf{z}} \quad (7)$$

$$\mathbf{F}_{\text{int}} = -[(\epsilon\text{Re}\{\chi\alpha_m^*\} + \mu\text{Re}\{\chi\alpha_e^*\})\sigma/n + \text{Re}\{\alpha_e\alpha_m^*\} + |\chi|^2]Ik^4/3\hat{\mathbf{z}} \quad (8)$$

Here $I = n^{-1}|\mathbf{E} \times \mathbf{H}^*| = |\mathbf{E}_0|^2$ is the intensity. The factor $\sigma = -2\text{Im}\{(\mathbf{E} \cdot \hat{\mathbf{x}})(\mathbf{E}^* \cdot \hat{\mathbf{y}})\}/(|\mathbf{E} \cdot \hat{\mathbf{x}}|^2 + |\mathbf{E} \cdot \hat{\mathbf{y}}|^2)$

is a measure for the degree of circular polarization (ellipticity) of the wave in the (x,y)-plane, such that $\sigma = +1$ for right circular polarization and $\sigma = -1$ for left circular polarization. In the calculations that follow, we will furthermore use the parameters $\tau = (|\mathbf{E} \cdot \hat{\mathbf{x}}|^2 - |\mathbf{E} \cdot \hat{\mathbf{y}}|^2)/(|\mathbf{E} \cdot \hat{\mathbf{x}}|^2 + |\mathbf{E} \cdot \hat{\mathbf{y}}|^2)$ and $\xi = 2\text{Re}\{(\mathbf{E} \cdot \hat{\mathbf{x}})(\mathbf{E}^* \cdot \hat{\mathbf{y}})\}/(|\mathbf{E} \cdot \hat{\mathbf{x}}|^2 + |\mathbf{E} \cdot \hat{\mathbf{y}}|^2)$ as measures for the linear polarization in the (x,y)-plane, such that $\tau = \{+1, -1\}$ correspond to linear polarization along $\{x, y\}$ and $\xi = \{+1, -1\}$ correspond to linear polarization at $\{+45^\circ, -45^\circ\}$ with respect to the x -axis. The helicity-dependent change of the radiation pressure due to the chiral polarizability χ in equation 8 has previously been used for sorting highly chiral liquid crystal droplets in optical lattices [8]. We now may consider an evanescent wave created by the total internal reflection of light at an interface in the (z,y)-plane, which has the form $\mathbf{E} = \sqrt{\mu}\mathbf{E}_0\exp(-\kappa x)\exp(ik_z z)$. If the evanescent field is propagating in a medium with index $n_2 = \sqrt{\epsilon\mu}$ and was created through the total internal reflection of a beam in a medium with index n_1 incident on the interface with angle θ in the (x,z)-plane (Fig. 1), then the wave vector components are given by $k_z = (n_1/n_2)\sin(\theta)k$, $k_x = \sqrt{k^2 - k_z^2}$ and $\kappa = \sqrt{k_z^2 - k^2} = -ik_x$, where $k = n_2\omega/c$. The expressions for all quantities relevant for the calculation of the forces with eqns. 2 and 3 are listed in the supplementary information [20]. Notably, evanescent fields have longitudinally polarized field components [20], which give rise to transverse spin angular momentum [13, 22]. In case of a p-polarized wave ($\tau = 1$) it is the electric field that is elliptically polarized in the (x,z)-plane, and in case of a s-polarized wave ($\tau = -1$) it is the magnetic field. Correspondingly, the out of plane electric and magnetic spin-components are given by $(\hat{\mathbf{y}} \cdot \mathbf{s}_e) = (1 + \tau)\frac{g}{4\omega}\frac{\kappa}{k_z}I_e$ and $(\hat{\mathbf{y}} \cdot \mathbf{s}_m) = (1 - \tau)\frac{g}{4\omega}\frac{\kappa}{k_z}I_e$ and the full expressions for the forces on a chiral dipole in an evanescent field are:

$$\mathbf{F}_0 = \kappa I_e [\text{Re}\{\chi\}n_2\sigma - \frac{1}{2}(\text{Re}\{\mu\alpha_e\}(1 + \tau\frac{\kappa^2}{k_z^2}) + \text{Re}\{\epsilon\alpha_m\}(1 - \tau\frac{\kappa^2}{k_z^2}))]\hat{\mathbf{x}} \quad (9)$$

$$\frac{I_e}{2}k_z[\text{Im}\{\mu\alpha_e\}(1 + \tau\frac{\kappa^2}{k_z^2}) + \text{Im}\{\epsilon\alpha_m\}(1 - \tau\frac{\kappa^2}{k_z^2}) + 2\text{Im}\{\chi\}\sigma n_2]\hat{\mathbf{z}}$$

$$\begin{aligned} \mathbf{F}_{\text{int}} = & -I_e\frac{k^4}{3}\frac{\kappa}{k_z}[\text{Re}\{\alpha_e\alpha_m^*\}\sigma + \text{Im}\{\alpha_e^*\alpha_m\}\xi + \sigma|\chi|^2 + \frac{\epsilon}{n_2}(1 - \tau)\text{Re}\{\chi\alpha_m^*\} + \frac{\mu}{n_2}(1 + \tau)\text{Re}\{\chi\alpha_e^*\}]\hat{\mathbf{y}} \\ & -I_e\frac{k^4}{3}\frac{k}{k_z}[\text{Re}\{\alpha_e\alpha_m^*\} + |\chi|^2 + \frac{\sigma}{n_2}(\epsilon\text{Re}\{\chi\alpha_m^*\} + \mu\text{Re}\{\chi\alpha_e^*\})]\hat{\mathbf{z}} \\ & -I_e\frac{k^4}{3}\frac{\kappa k}{k_z^2}(\frac{\xi}{n_2}(\text{Re}\{\mu\chi\alpha_e^*\} - \text{Re}\{\epsilon\chi\alpha_m^*\}) + \text{Im}\{\alpha_e^*\alpha_m\}\tau)\hat{\mathbf{x}} \end{aligned} \quad (10)$$

with $I_e = \frac{|\mathbf{E}_0|^2}{1+\tau(\kappa/k_z)^2} \exp(-2\kappa x)$. The lateral forces experienced by the dipole are given by the first line of eqn. 10, with all chirality-dependent terms proportional to χ . Unfortunately the chirality-dependent lateral forces caused by $\nabla \times \mathbf{p}$ and \mathbf{s} in for the \mathbf{F}_0 force exactly cancel each other [23]. The y -component of the Poynting momentum density \mathbf{p} arises entirely from the Belifante spin momentum density \mathbf{p}^s for polarizations with non-vanishing σ , which also gives rise to a helicity-independent lateral force term on chiral particles. An analogous y -term arises as part of the imaginary Poynting momentum density \mathbf{p}'' for polarizations with non-vanishing ξ . The intensity and polarization of the evanescent wave depend on the complex transmission coefficients for the totally internally reflected beam.

To illustrate characteristic magnitudes of the force, the strength of the lateral force acting on a spherical nanoparticle is shown in Fig 2 as a function of chirality and particle radius in direct comparison with the lateral forces that arise due to the mechanisms described by Wang et. al [12] and Bliokh et. al [13]. In each case, the lateral force emerging through the interaction of the transverse SAM with the chiral particle is stronger by about 1.75 and four orders of magnitude over the considered range. Here material chirality is parametrized with a chirality parameter $K \in [-1, 1]$ as in ref. [12], and the derivation of the expression of the force on such a sphere is listed in the supplementary information [20], as well as a discussion of the force on helicene molecules and gold nanohelices, which represent other types of chiral particles frequently considered in the literature.

CONCLUSION

We predict lateral forces on materials with chiral optical response in evanescent fields, which push particles with opposite helicities in opposite directions with strength dependent on the chiral polarizability. The forces result from the interaction of the evanescent field's transverse optical SAM density of the wave with the chiral electromagnetic response of the particle, and are particularly strong in comparison to previously predicted lateral optical forces. Transverse SAM may arise whenever light is laterally confined, so that the effect described in this paper represents a natural choice for the optical sorting by material chirality in an integrated system. The effect can be produced by a single beam, which significantly aids alignment and avoids standing wave patterns that limit the separation of enantiomers to the width of interference fringes. However, the use of multiple beams may enable the cancellation of the longitudinal force component or, given the possibility of generating transverse spin, even the generation of spin-optical lateral forces in free space beams [25]. While we limited the discussion to the simplest case of an evanescent field created by

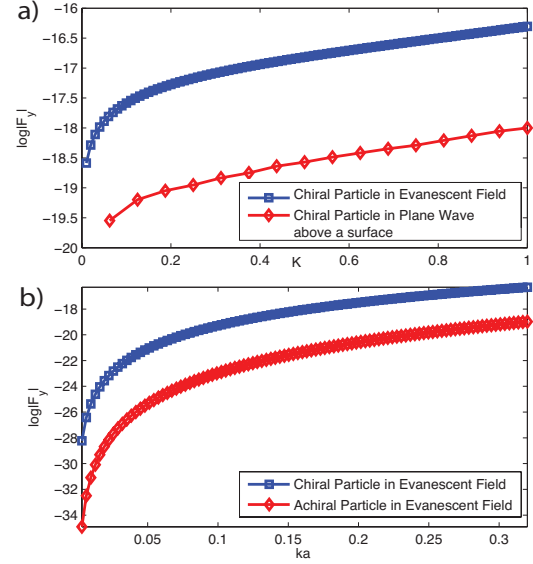


FIG. 2. Lateral Optical Forces: The previously predicted lateral optical forces in comparison to the lateral force on a chiral particle in an evanescent field. **a)** The lateral optical force on a chiral nano sphere in a plane wave above a reflective surface according to Wang et. al. [12] and due to an evanescent field in $\text{N}/(\text{mW}/\mu\text{m}^2)$ as a function of the chirality parameter K [20, 24]. The sphere is non-magnetic, situated 60nm above the surface and has a dielectric constant of $\epsilon_s = 2$ and a radius of 30nm. In case of the force in the evanescent field, the intensity refers to the intensity of a beam that is totally internally reflected at a flint glass/air interface at an angle of $\theta = 36.4^\circ$. **b)** The lateral force on an achiral sphere ($K = 0$) due to the Belifante spin momentum density according to Bliokh et. al. and on an equivalent chiral sphere ($K = 1$) as a function of ka , where a is the radius of the sphere and $k = n_2\omega/c$ is the wavenumber of the evanescent field. In both cases the sphere is non-magnetic and situated at a height of 30nm, has dielectric constant $\epsilon_s = 2$ and experiences the evanescent field generated at a flint glass/air interface at an incident angle of $\theta = 36.4^\circ$.

the total internal reflection of light at a single interface, there is significant potential for improving the strength of the force by further engineering the light field. Natural choices for this are the intense and inherently evanescent fields of plasmonic excitations, or optical waveguides designed to have regions with high field enhancement, such as slot waveguides [26]. We furthermore limited the discussion to the Rayleigh limit, where compact closed form expressions exist, though the optical manipulation of objects smaller than a few 100 nm in liquid suspension is in general very challenging due to thermal agitation. Other than by tailoring the light fields, this may in particular be overcome in a low pressure environment. Larger particles may take advantage of resonances (such as Mie resonances) and are bound to experience much stronger forces.

ACKNOWLEDGEMENTS

This research was supported by the Airforce Office of Scientific Research (AFOSR) under Grant no. FA9550-12-1-0289. The authors thank X. Yin for helpful discussions.

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